

(12) **UK Patent Application** (19) **GB** (11) **2 320 503** (13) **A**

(43) Date of A Publication **24.06.1998**

(21) Application No **9626654.9**

(22) Date of Filing **21.12.1996**

(71) Applicant(s)

**Pilkington Plc**

**(Incorporated in the United Kingdom)**

**Prescot Road, ST HELENS, Merseyside, WA10 3TT,  
United Kingdom**

**Libbey Owens-Ford Company  
811 Madison Avenue, Toledo, Ohio 43695,  
United States of America**

(72) Inventor(s)

**Anthony Alan Parker  
Julia Bronwen MacLachlan  
Neil Winstanley  
Paul Arthur Holmes**

(74) continued overleaf

(51) INT CL<sup>6</sup>

**B32B 17/10 , C08L 27/06**

(52) UK CL (Edition P )

**C3V VAM VEF**

**B5N N1710 N195 N196 N200 N207 N21Y N214 N223  
N255 N257 N273 N2730 N28Y N295 N297 N30Y N33X  
N33Y N331 N337 N339 N34X N342 N343 N345 N349  
N35X N361 N362 N363 N46X N463 N474 N478 N479  
N491 N57Y N575 N595 N596 N597 N611 N648 N650  
N658 N66Y N661 N670 N672 N734 N753 N754 N771  
C3M MXC M110 M153 M155 M156 M158 M170  
C3W W217 W221 W225 W302 W319  
U1S S1386 S1591 S1820 S1839**

(56) Documents Cited

**WO 95/19261 A1 US 4277538 A**

(58) Field of Search

**UK CL (Edition O ) C3M MXC , C3V VAM VBA VEF  
VEG  
INT CL<sup>6</sup> B32B  
Online: WPI**

(54) **Safety glass interlayer**

(57) A self-adhering composite interlayer formed from a plasticized polyvinyl chloride containing film, a silane and a heat stabilizer, the interlayer having a pummel value of at least 3, a yellowing index of less than 5 and a haze of less than 1 and laminated safety glass in which such a interlayer is sandwiched between two glass sheets. The film may comprise copolymers of vinyl chloride with ethylene, vinyl acetate or alkyl acrylate and polycaprolactone.

**GB 2 320 503 A**

(74) Agent and/or Address for Service

**Michael John Lee**

**Pilkington Plc, Group Patents Department, Pilkington  
Technology Centre, Hall Lane, Lathom, ORMSKIRK,  
Lancashire, L40 5UF, United Kingdom**

## **SELF-ADHERING POLYVINYL CHLORIDE SAFETY GLASS INTERLAYER**

### **BACKGROUND OF THE INVENTION**

#### **1. Field of the Invention**

The present invention relates to an interlayer for safety glass and, more particularly, to a self-adhering interlayer including a polyvinyl chloride polymeric material.

#### **2. Summary of Related Art**

Safety glass is a well known term for a glass sandwich composed of an interlayer bonding together two glass plates or sheets so that breaking the glass results in minimum dispersion of broken glass fragments. The interlayer must possess a number of properties, including the following: high impact energy adsorption to minimize concussive injury; shear and tear strength sufficient to prevent rupture of the interlayer by the broken glass; sufficient adhesion to the glass to inhibit laceration on contact with, and prevent dispersion of, broken glass; acceptable thermal heat stability and weatherability; and good optical quality. The interlayer must possess these properties over the wide range of temperatures in which these laminated glazings are used.

It has been widely known to use a film of plasticized polyvinyl butyral as the interlayer material in safety glass for automobiles, air planes and building materials because of its high adhesiveness, transparency and good mechanical properties over a broad range of temperatures. However, the use of plasticized polyvinyl butyral films makes the production of laminated safety glass relatively expensive.

The surface of plasticized polyvinyl butyral film is very tacky, and presents a problem of blocking at the time of windup after film formation. Thus, plasticized polyvinyl butyral film must be provided with some parting means if it is to be stored or transported in the form of stacks of die-cut blanks or in the form of rolls. Furthermore, the production of plasticized polyvinyl butyral films requires specialised equipment and, due to their sensitivity to moisture, plasticized polyvinyl butyral films must generally be handled under controlled atmosphere conditions during manufacture, storage and immediately prior to their incorporation into the laminated safety glass. This all adds to the expense of utilizing plasticized polyvinyl butyral films in laminated safety glass.

Alternative interlayer materials have been proposed. For instance, US Patent No 4,277,538 to Beckmann et al discloses a laminated safety glass employing a sheet of plasticized polyvinyl chloride (PVC) as the interlayer. The use of PVC would be advantageous in that it may be produced on conventional equipment and would be much less expensive to manufacture and process into a laminated safety glass when compared with polyvinyl butyral. However, by itself, a PVC film will not adhere to glass. To increase the adhesion of the PVC to glass, Beckmann et al suggest the use of an organofunctional silane, either as a primer or uniformly dispersed within the PVC film.

However, neither use of a silane adhesion promoter is without drawbacks. Dispersing an organofunctional silane within the PVC film in quantities sufficient to provide adequate adhesion to glass has hitherto been found to result in a laminate having a haze which is unacceptably high and an unacceptable heat stability which makes the film too yellow for many applications. In addition, dispersing the silane in the interlayer may have a negative effect on the processability of the interlayer material. Secondly, the application of an organofunctional

silane adhesion promoter as a primer necessitates an additional lamination manufacturing step, increasing the manufacturing costs of the resulting laminated glazing unit.

US Patent No 4,600,627 discloses a laminated glazing unit including a pair of glass sheets and a composite interlayer. The interlayer is formed of an interleaving sheet formed from polyester or PVC sandwiched between two intermediate layers of a crosslinking polymer, such as ethylene/vinyl acetate (EVA). The intermediate layers, which provide an adhesive bond between the interleaving sheet and the two glass sheets, have a thickness of about 0.2 mm. It has been determined, however, that intermediate layers of EVA will not provide sufficient adhesion to the glass, and that the use of silane adhesion promoters is therefore required. In addition, at least where the interleaving sheet is formed of PVC, the resulting laminating glazing exhibits poor optical quality.

Accordingly, the present invention seeks to provide an improved PVC containing interlayer which is relatively inexpensive and self-adhering to a glass sheet. Furthermore, the present invention seeks to provide such a PVC containing interlayer which, when incorporated into a laminated glazing, provides good optical quality.

#### SUMMARY OF THE INVENTION

The present invention relates to an improved polyvinyl chloride containing interlayer for laminated safety glass. The present invention also relates to laminated safety glass or glazing unit including a pair of glass sheets bonded together with a self-adhering interlayer.

According to the present invention, there is provided a self-adhesive polyvinyl chloride-containing interlayer composition comprising a plasticized polymer of vinyl chloride having incorporated therein a silane and a primary heat stabilizer, the interlayer composition having a pummel value of at least 3, a yellowing index of less than 5 and a haze value of less than 1. Preferably, the silane selected is an aromatic secondary aminosilane. Advantageously, the

primary heat stabilizer comprises at least one organometallic salt, the metal being selected from the group consisting of aluminium, barium, calcium, cadmium, lead, magnesium, tin and zinc.

In the prior art, particularly in United States Patent Specification No 4,277,538, there are disclosed an extremely large number of possible interlayer compositions suitable for forming a laminated safety glass. Such prior specification also states that the silanes may be added to the molding compositions as they are. It is presumed that this means that the silane may be incorporated directly in the polyvinyl chloride-containing polymeric material.

This somewhat bland statement disguises the actual situation in practice. For an interlayer to be suitable for use in a laminated safety glass, it must satisfy a number of criteria. Firstly, the adhesive must be capable of adhering to glass. However, the interlayer must be in the form of a film and this latter is produced mechanically. Such mechanical treatment is, generally, milling and/or calendering. If the adhesion of the adhesive is too great, the interlayer adheres to the mill and/or calendar and it is not possible to produce the film. If it is too weak, it adheres insufficiently to the glass.

Secondly, because the interlayer is intended for use in glazing, it must have good optical properties. In practice, this means that it must have a low yellowing factor and a low haze factor, and rheological properties that enable processing as a flat sheet to minimize optical distortion. Yellowing is a major problem in most plastics materials and, in the case of laminated safety glasses of the present type, is caused by the thermal degradation of the adhesive and of the polymer. Haze is caused by phase separation and, clearly, is more likely to occur when one attempts to incorporate the adhesive into the polyvinyl chloride than if the former is simply applied as a coating to the latter. The thickness profile of sheets can be linked to the melt flow rheological characteristics of the formulation and can be greatly altered by chemical crosslinking reactions with adhesive components that are added to the

formulation. These problems are somewhat interlinked. For example, as the temperature is increased, polyvinyl chloride resins turn more yellow. To prevent this, heat stabilizers are added to the resin. However, this, in turn, increases the number of phases present and tends to lead to increased haze, and can also lead to higher melt viscosity, which can adversely affect the thickness profile of the sheet..

For this reason, although the above-identified United States Patent was published in 1981, we are not aware of any laminated glazing units having a polyvinyl chloride-containing polymeric interlayer in which the adhesive has been incorporated. In fact, the most recent prior art, such as European Patent Specification No EP 0603073A1, has concentrated on providing a laminated safety glass in which the adhesive and the polyvinyl chloride-containing polymer are kept separate from one another. In other words, the glass comprises a glass-adhesive-polyvinyl chloride-adhesive-glass sandwich. However, this is not particularly desirable because it introduces a further stage into the manufacture of the glazing. Incorporation, on the other hand, does not.

We have surprisingly found that it is possible to incorporate an adhesive into a polyvinyl chloride-containing polymer to form an interlayer which has the necessary optical properties. We have found that this is dependent upon the careful selection of a silane as the adhesive agent, the particular primary heat stabilizer employed and the polymer in which the silane is to be incorporated.

As stated above, the preferred silane employed is an aromatic secondary aminosilane. Although the mechanism of the bonding is not fully understood, this type of silane includes a heteroatom having a lone pair and it is believed that this has a modifying effect on the alkoxy groups in the silane which become hydrolysed to form hydroxyl groups which, in turn, bond to the glass.

The efficacy of an aromatic silane is totally unexpected. Normally, it would not be expected that an aromatic silane would have any advantages over an aliphatic silane and would have a major disadvantage. We have attempted to incorporate aliphatic silanes into a polyvinyl chloride-containing polymer and have encountered two major problems. Firstly, the adhesion is too high and the film cannot be processed and, secondly, the yellowing index is extremely high. One would normally expect this second problem to be exacerbated if an aromatic silane were used. As mentioned hereinbefore, yellowing is caused by thermal degradation and aromatic degradation products are, in general, more yellow than their aliphatic counterparts. We have, however, found that an aromatic secondary aminosilane, particularly N-(3-trimethoxysilyl) propyl benzenamine, has more controlled adhesion and has a low yellowing factor.

Even if it is possible to incorporate such a silane, it is still necessary to find an appropriate heat stabilizer which does not adversely affect the yellowing factor or the haze. Although the heat stabilizers which are organometallic salts are known as a class, we have found that only a mixture of calcium or barium salts with zinc salts, preferably the sebacates, produces the desired results. Furthermore, we have determined that the polymer in which the aromatic silane and the heat stabilizer are incorporated is, advantageously, a blend of a vinyl chloride homopolymer with a vinyl chloride-ethylene copolymer.

It is our belief that it is the combination of the silane, the primary heat stabilizer and the polyvinyl chloride-containing polymer which makes it possible to produce an interlayer in which the former two components are incorporated in the latter.

#### DESCRIPTION OF THE PREFERRED EMBODIMENTS

The numerous and varied properties required of an interlayer material for use in laminated safety glass had made it very difficult to successfully develop such laminated safety

glass employing a PVC interlayer, even though the cost advantages of so doing are potentially substantial. As mentioned above, the properties an interlayer must possess include: high impact energy adsorption; shear and tear strength sufficient to prevent rupture of the interlayer by broken glass; sufficient adhesion to the glass to inhibit laceration on contact with, and prevent dispersion of, broken glass; acceptable thermal heat stability and weatherability; and good optical quality.

The polyvinyl chloride-containing polymer of the interlayer composition is formed of a relatively high number average molecular weight vinyl chloride polymer. Thus, it may be in the form of a vinyl chloride homopolymer, copolymer or graft polymer, or blended combinations thereof. The thickness of the interlayer substantially affects the impact strength of the laminated glazing unit. The impact strength of the laminated glazing unit will decrease with a decrease in the thickness of the interlayer so the preferred thickness is at least 25 mils. However, from a commercial standpoint, it is desirable to form the interlayer so that the overall thickness and weight of the laminated glazing unit is comparable to current products, which utilize a PVB interlayer about 30 mils thick. Accordingly, the thickness of the interlayer is preferably between about 28 to 35 mils.

In a particularly preferred embodiment, the polymer is formed from a blend of a homopolymer of polyvinyl chloride such as SE-950 having a degree of polymerisation of 950 and available from Shintech Inc of the United States with an ethylene-vinyl chloride copolymer such as VE-U resin, commercially available from Sekisui of Japan. This poly(vinyl chloride-co-ethylene) includes 16% ethylene comonomer.

If the interlayer is formed of a polyvinyl chloride homopolymer, it preferably has a number average molecular weight, or a mixture of number average molecular weights, of at least 30,000. In a preferred embodiment, the number average molecular weight, or mixture of

number average molecular weights, is between about 40,000 and about 165,000, with a degree of polymerization of about 500 to 2600. One such preferred PVC resin is SE-950, having a degree of polymerization of 950 which is commercially available from Shintech Inc of Houston, Texas.

Control of the molecular weight distribution and purity of the PVC has been found to be very important in a safety glass interlayer application. Variances in molecular weight distribution can have a profound effect on the impact properties and processability of the film. In addition, the presence of impurities, such as initiators and surfactants, can lead to excessive degradation and yellowing and/or haze of the interlayer and can have detrimental effect on the long term heat stability of the interlayer. Accordingly, the PVC is preferably formed by suspension polymerization, which offers superior control of molecular weight with the lowest level of residual impurities.

It has also been found that a plasticized film containing a blend of a PVC homopolymer with copolymers, such as a copolymer between vinyl chloride and vinyl acetate, provides an improved interlayer. Such a copolymer exhibits improved flow properties, and hence, when blended with the PVC homopolymer results in improved processability over the PVC homopolymer alone. The polymer may, therefore, be composed of a blend of from about 75 to 98 phr PVC and from about 2 to 25 phr poly(vinyl chloride-co-vinyl acetate). In a preferred polymer of this type, the blend is about 95 parts PVC homopolymer and about 5 parts of the poly(vinyl chloride-co-vinyl acetate). Examples of suitable vinyl chloride/vinyl acetate copolymers are MPR-TSN, commercially available from Nissin Chemicals, Nitta-Gun, Japan, which is a copolymer of 87% vinyl chloride and 13% vinyl acetate with a degree of polymerization of 400; and UCAR VYHD, commercially available from Union Carbide, which

is a copolymer of 86% vinyl chloride and 14% vinyl acetate with a degree of polymerization of 220.

In another preferred embodiment, the interlayer 12 is formed of a blend of a PVC homopolymer and a copolymer of vinyl chloride and methylacrylate. An example of a suitable vinyl chloride/methylacrylate copolymer is Geon E8 commercially available from The Geon Company. In this embodiment, the polymer 12 is formed from about 40 to 80 parts PVC homopolymer with about 20 to 60 parts of the vinyl chloride/methylacrylate copolymer. Any of the blends described above may also preferably include from about 5 to 15 parts of a polycaprolactone, such as CAPA 656, commercially available from Solvay Interlox.

The plasticizers for the preparation of the plasticized films containing PVC in accordance with the invention can be linear or branched aliphatic diesters, triesters or tetraesters, or aromatic diesters, triesters or tetraesters, or mixtures thereof. From a performance standpoint, preferred plasticizers include dihexyl azelate (DHZ), dihexyl adipate (DHA) and dioctyl azelate (DOZ). However, dioctyl adipate (DOA), while providing slightly reduced performance relative to the other noted plasticizers, may be most preferred in view of its combination of performance and relatively lower cost. In addition, plasticizer blends may be employed for both economic and performance considerations. For example, mixing dioctyl sebacate (DOS) or DOP with DHA will result in a higher glass transition temperature than with DHA alone at the same total plasticizer level. As a result, room temperature mean break height will improve without disrupting low temperature performance. The total plasticizer concentration is between about 20 to about 60 phr, depending primarily upon the number average molecular weight and molecular weight distribution of the PVC containing resin. A total plasticizer concentration of between about 30 and about 45 phr is most preferred.

The PVC containing film has incorporated therein a primary heat stabilizer which is advantageously an organometallic compound, such as a salt of an alkaline earth metal or selected transition metals, including aluminium, barium, cadmium, calcium, lead, magnesium, tin, and zinc. The heat stabilizer is desirably present in an amount of between 1 and 5, optimally 2 and 4 phr. If the silane is an aromatic secondary aminosilane, the primary heat stabilizer preferably includes a mixture of a zinc salt of an organic acid and a calcium salt of an organic acid. The primary heat stabilizer preferably includes from about 1.6 to 4.0 % atomic zinc as a zinc salt of an organic acid, and from about 7.0 to 14.0 % atomic calcium as a calcium salt of an organic acid. The metals salts may be, for example, stearates, sebacates, laurates, oleates, iso-stearates, octoates, decanates, nonylphenoates or mixtures thereof.

To maximise the long term, service use temperature heat stability of a PVC film used as a safety glass interlayer, the primary heat stabilizer also preferably includes from about 2.0 to 4.0 % phosphorus as phosphites. A preferred phosphite has been found to be triphenyl phosphite.

In addition to the primary heat stabilizer, the PVC containing film also preferably includes one or more secondary heat stabilizers, including epoxidised oils, perchlorates, 1,3 [beta]-diketones and mixtures thereof. From about 2.5 to 15.0 phr of an epoxidised oil is preferably included as a secondary heat stabilizer in the PVC containing film. A preferred epoxidised oil is epoxidised soybean oil. The PVC film also preferably includes from about 0.1 to 1.0 phr of a perchlorate, a preferred perchlorate being sodium perchlorate. As another secondary heat stabilizer, the PVC film may be provided with between about 0.1 and 2.0 phr of a 1,3 [beta]-diketone. A preferred 1,3 [beta]-diketone has been found to be stearyl benzoyl methane.

The polymer may also include other additives, such as UV light stabilizers, antioxidants, optical brighteners, dyes and the like. Thus, the polymer is preferably formed from a

formulation including from about 0 to 2 phr of a benzophenone or benzotriazole derivative as a UV stabilizer and from about 0 to 5 phr hindered phenols as an antioxidant. From about 0 to 1 phr of a fluorescent or whitening agent and from about 0 to 1 phr of a blue dye may also be incorporated in the polymer if desired.

The adhesive of the interlayer composition is a silane which can be incorporated in the polymer and which adheres to glass sheets.

As noted above, the adhesive must be incorporatable in the polymer as well as adhering to the glass sheets. To accomplish this, the adhesive is formed of a material which is compatible with the vinyl chloride polymer and includes at least one type of functional group which is capable of forming chemical bonds with glass, whether such bonds be covalent, ionic, hydrogen bonds, or other chemical bonds.

In a preferred embodiment, the adhesive is a silane which both bonds to the glass and remains dispersed in the PVC - either by entanglement or, more likely, chemical bonding. Aromatic secondary aminosilanes are preferred.

### EXAMPLES

The following examples are illustrative of the present invention and do not constitute any limitation with regard to the subject matter of the invention. The adhesion, optical quality, and heat stability of each of the laminates in these examples were measured by the following methods.

#### 1. Adhesion

The pummel test was used to measure interlayer adhesion to the glass. Twelve inch square glass laminates were placed in a -18°C refrigerator for at least two hours. After removal from the refrigerator, the laminates were placed on a metal substrate and hit

repeatedly with a 16 oz hammer to break the glass. All broken glass unadhered to the interlayer was then removed. The amount of glass left adhered to the interlayer was visually compared with a set of standards of known pummel scale and a pummel value for each sample was assigned, ranging from a pummel value of 0 (no adhesion; no glass adhered) to 10 (high adhesion, 100% of the glass adhered).

## 2. Optical Quality

The optical quality of laminated samples was determined by measurement of transmitted distortion using a float glass distortion meter, and by visual inspection using both a shadowgraph and a gridboard background. Transmitted haze was measured with a Hazegard XL200 from Gardner/BYK.

## 3. Heat Stability

Heat stability was determined by monitoring the yellowing rate of four inch square laminated samples in one or more temperature controlled ovens. After measuring the initial yellowness index (YIC) using the Spectrogard from Gardner/BYK of Silver Springs, Maryland, samples were placed in one of five ovens set at various temperatures (65, 80, 100, 120 and 150°C). The samples were removed from the ovens at regular intervals and the YIC was measured. The various time intervals were as follows: 500 hours for the oven at 65°C, 250 hours for the oven at 80°C, 48 hours for the oven at 100°C, 24 hours for the oven at 120°C, and 4 hours for the oven at 150°C.

## EXAMPLE 1

An interlayer approximately 0.8 mm (30 to 33 mils) thick was made having the following formulation:

<u>Component</u>	<u>phr</u>
SE950 PVC Resin <sup>1</sup>	60
VE-U <sup>2</sup>	30
CAPA 650 <sup>3</sup>	10
Dihexyl adipate	12.8
Dioctyl sebacate	19.2
Baerlocher TD60-2 <sup>4</sup>	3
Drapex 6.8 <sup>5</sup>	5
CPL 46 <sup>7</sup>	0.1
Tinuvin 328 <sup>8</sup>	0.75
Y-9669 <sup>9</sup>	1

1. PVC resin having a degree of polymerization of 950 available from Shintech Inc of Freeport, TX.
2. Copolymer resin (84 mol % vinyl chloride - 16% ethylene monomermixture) available from Sekisui Fire Chemical Co Ltd of Osaka, Japan.
3. Polycaprolactone resin available from Solvay Interlox.
4. Calcium/zinc stabilizer package available from Baerlocher GmbH of Munich, Germany.
5. Epoxidised soybean oil available from Witco Corp. of Oakland, NJ.
6. Hindered phenolic antioxidant available from Ciba-Geigy Corp. of Hawthorn, NY.
7. Perchlorate stabilizer available from Asahi Denka Kogyo K.K. of Japan.
8. Benzotriazole ultraviolet light stabilizer available from Ciba-Geigy Corp.
9. N-(3-trimethoxysilyl) propyl benzenamine available from Union Carbide Corporation.

The above interlayer is produced by mixing the components in, initially a high speed mixer and then in an intensive, shear-type, mixer. The mixers are both heated so that the temperature of the interlayer rises to approximately 160°C. The interlayer is then subjected to the action of two sequential two-roll mills and is thereafter calendered. Laminates are then

produced by applying glass sheets to opposed sides of the interlayer and are autoclaved at approximately 150°C and 240 psi with a 45 minute hold time.

Clear laminates were obtained with the following results:

<u>Property</u>	<u>Value</u>
Adhesion, initial (pummel)	9
Haze, initial	0.8%
Heat stability (yellowing index)	1.19

## EXAMPLE 2

An interlayer composition was produced as described in Example 1 with the exceptions that the calcium/zinc stabilizer was replaced by the same amount of a barium/zinc stabilizer (UBZ 791 made by Baerlocker USA of Dover, Ohio) and that 1.0 phr of the silane was used.

The test values obtained were as follows:

<u>Property</u>	
Adhesion, initial (pummel)	9
Haze, initial	0.5
Yellowing index	0.95

However, we have found that the initial adhesion (pummel value) is dependent upon the hydrolysis of the silane. Under very dry conditions, such hydrolysis may only proceed slowly. If a film is produced which has no adhesion or an adhesion which is too low, this can be rectified by effecting a further treatment. This further treatment may be effected by subjecting the film alone to conditioning at substantially ambient temperatures in an environment having a high relative humidity. For example, such conditioning is usually effected for a period of at least 24 hours with a relative humidity of approximately 70%.

Alternatively, the further treatment comprises subjecting the interlayer assembly to heat treatment at an elevated temperature. For example, heating a sample to 110°C for a period of 18 hours will typically increase the pummel value from about 3-4 to about 7-8. Although we do not currently consider both of these treatments to be necessary, the possibility of so doing has not been discounted.

There is, on occasion, a tendency for an interlayer made of the material of the present invention to "age" too quickly. This manifests itself by an increase in the yellowing index. We have, however, found that this problem can be obviated, or at least minimised, by sandwiching the film between two layers of glass, at least one of which is tinted and has good ultraviolet absorption properties. The tinted glass may include a colorant selected from the group consisting of iron, cerium and titanium oxides and mixtures thereof. The ultraviolet transmission of the glass, over the range of 300 nm to 400 nm is preferably below 68%, desirably below 56%, at a thickness of 2.1 mm.

**Claims**

1. A self-adhesive polyvinyl chloride-containing interlayer composition comprising a plasticized polymer of vinyl chloride having incorporated therein a silane and a primary heat stabilizer, the interlayer composition, when formed as a film having a nominal thickness of 30 mils and laminated between a pair of glass sheets, having a pummel value of at least 3, a yellowing index of less than 5 and a haze value of less than 1.
2. An interlayer composition as claimed in claim 1 wherein the silane is a secondary aminosilane.
3. An interlayer composition as claimed in claim 2 wherein the silane is an aromatic secondary aminosilane.
4. An interlayer composition as claimed in claim 1 or 2 wherein the primary heat stabilizer comprises at least one organometallic salt, the metal being selected from the group consisting of aluminium, barium, calcium, cadmium, lead, magnesium, tin and zinc.
5. An interlayer composition as claimed in claim 2 or 3 wherein the silane is N-(3-trimethoxysilyl) propyl benzenamine.
6. An interlayer composition as claimed in claim 4 wherein the primary heat stabilizer is a mixture of a calcium salt or a barium salt of an organic acid and a zinc salt of an organic acid.
7. An interlayer composition as claimed in claim 1, 3, 4 or 5 wherein the organometallic salt is selected from the group consisting of stearates, laurates, oleates, isostearates, octoates, decanoates and nonylphenolates and mixtures thereof.

8. An interlayer composition as claimed in any preceding claim wherein the polymer of vinyl chloride is selected from the group consisting of vinyl chloride homopolymers, vinyl chloride copolymers, vinyl chloride graft polymers and blended combinations thereof.
9. An interlayer composition as claimed in claim 10 wherein the polymer is selected from the group consisting of a blend of vinyl chloride homopolymers and co-polymers of vinyl chloride with a polymerisable monomer selected from the group consisting of lower alkenes containing from 1 to 4 carbon atoms, vinyl acetate, and lower alkyl acrylates wherein the lower alkyl group contains from 1 to 4 carbon atoms.
10. An interlayer composition as claimed in claim 8 wherein the polymer is a blend of a homopolymer of vinyl chloride with a co-polymer of vinyl chloride and ethylene.
11. An interlayer composition as claimed in any preceding claim wherein the plasticizer is selected from the group consisting of aliphatic diesters, aliphatic triesters, aliphatic tetraesters, aromatic diesters, aromatic triesters and mixtures thereof.
12. An interlayer composition as claimed in claim 10 wherein the esters are selected from the group consisting of azelates, adipates, sebacates and phthalates and mixtures thereof.
13. An interlayer composition as claimed in claim 11 wherein the plasticizer comprises a mixture of dihexyl adipate and dioctyl sebacate.
14. An interlayer composition as claimed in any one of claims 10 to 12 wherein the plasticizer further includes a polycaprolactone.
15. An interlayer composition as claimed in any preceding claim additionally including a secondary heat stabilizer.
16. An interlayer composition as claimed in claim 14 wherein the secondary heat stabilizer is a mixture of epoxidised soyabean oil and perchlorate.

17. An interlayer composition as claimed in claim 15 wherein the perchlorate is sodium perchlorate.
18. An interlayer composition as claimed in any preceding claim additionally including an antioxidant.
19. An interlayer composition as claimed in claim 17 wherein the antioxidant is a hindered phenolic antioxidant.
20. An interlayer composition as claimed in any preceding claim additionally including an ultraviolet radiation absorber.
21. An interlayer composition as claimed in claim 19 wherein the absorber is selected from the group consisting of benzophenones and benzotriazoles.
22. An interlayer composition comprising a blend of a homopolymer of vinyl chloride with a co-polymer of vinyl chloride and ethylene; N-(3-trimethoxysilyl) propyl benzenamine; a primary heat stabilizer comprising a mixture of calcium and zinc stearates; a plasticizer comprising a mixture of dihexyl adipate and dioctyl sebacate; a secondary heat stabilizer comprising a mixture of epoxidised soyabean oil and sodium perchlorate, a benzotriazole ultraviolet radiation absorber, a caprolactone; and a hindered phenolic antioxidant.
23. A laminated glass comprising a first glass sheet, a composition as claimed in any one of claims 1 to 21 in the form of a film adhering directly to said first glass sheet and a second glass sheet to which the film adheres directly.
24. A laminated glass as claimed in claim 23 wherein at least one of said glass sheets is tinted and contains at least one colorant which absorbs ultraviolet radiation.
25. A laminated glass as claimed in claim 24 wherein the colorant is selected from the group consisting of iron, cerium and titanium oxides and mixtures thereof.

26. A laminated glass as claimed in claim 25 wherein the tinted glass has an ultraviolet transmission of less than 68% over the range of 300 nm to 400 nm at a thickness of 2.1 mm.
27. A laminated glass as claimed in claim 26 wherein the tinted glass has an ultraviolet transmission of less than 56% over the range of 300 nm to 400 nm at a thickness of 2.1 mm.
28. An interlayer composition as claimed in claim 1 substantially as hereinbefore described with reference to the foregoing Examples.
29. A laminated glass as claimed in claim 23 substantially as hereinbefore described.



20

Application No: GB 9626654.9  
Claims searched: 1-29

Examiner: K. Macdonald  
Date of search: 12 March 1997

**Patents Act 1977**  
**Search Report under Section 17**

**Databases searched:**

UK Patent Office collections, including GB, EP, WO & US patent specifications, in:  
UK Cl (Ed.O): C3V(VAM,VBA,VEF,VEG); C3M(MXC)  
Int Cl (Ed.6): B32B  
Other: Online: WPI

**Documents considered to be relevant:**

Category	Identity of document and relevant passage	Relevant to claims
X	WO 95/19261 A1 (LIBBEY-OWENS-FORD) see Claim 1; page 8, lines 15-25; page 9, lines 17-22; page 12, lines 5-19; page 30, lines 9-17	1 at least
X	US 4277538 (DYNAMIT) see Claim 1; column 3, lines 42-50; column 5, lines 21-37	1 at least

X	Document indicating lack of novelty or inventive step	A	Document indicating technological background and/or state of the art.
Y	Document indicating lack of inventive step if combined with one or more other documents of same category.	P	Document published on or after the declared priority date but before the filing date of this invention.
&	Member of the same patent family	E	Patent document published on or after, but with priority date earlier than, the filing date of this application.